

PATENT SPECIFICATION

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(54) HOT MELT COMPOSITION

(71) We, TOYO SODA MANUFACTURING CO. LTD., a Japanese company of No. 4560, Oaza-tonda, Shin-nanyo-shi, Yamaguchi-ken, Japan, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to hot melt compositions and to a method of raising the softening temperature of a hot melt composition containing an ethylene-vinyl acetate copolymer and/or a rubber as a main component.

In general, conventional hot melt compositions have been prepared by blending a material mainly consisting of an ethylene-vinyl acetate copolymer and at least one additive selected from tackifiers, waxes and asphalts, and if desirable, rubbers, plasticizers, non-adhesive resins, pigments or fillers. The compositions are solid at room temperature and are used by melting and re-solidifying. Various hot melt type compositions have been prepared and may be classified as follows:

1) Hot melt adhesive compositions

The compositions are adhesives which are solid at room temperature and are applied in a heat molten condition. The compositions bond two substrates by cooling below the melting point. The hot melt adhesive compositions are prepared by blending, melting and mixing a material mainly consisting of an ethylene-vinyl acetate copolymer, for imparting adhesive and cohesive properties, with:

a tackifier, for improving adhesive properties and processability;
 a wax, for decreasing viscosity in the melt condition and preventing blocking, and, if desired, a plasticizer, for imparting bendability;
 a rubber;

an antioxidant, for preventing oxidation or deterioration at high temperature;
 a pigment, for coloring;

or other additives.

The hot melt adhesive compositions can be used in various fields, such as bookbinding,

woodworking, plywood making, packaging, canning, shoe making, automatic processing, manufacture of paper articles, building, electrical parts and vehicle parts. The hot melt adhesive compositions are prepared by selecting a formulation which will achieve the desired adhesive properties, melt viscosity, thermal stability, bendability, heat resistance, cold resistance, threadability and blocking properties of the composition.

2) Hot melt coating compositions

The properties required in a hot melt coating depend upon the intended usage thereof, and include heat resistance, heat rolling ability, lustre, moistureproofness, softness, antiblocking and transparency. The compositions must have low viscosity in the melt condition, since the compositions are applied in a molten condition onto a substrate. The hot melt coating compositions are usually composed of an ethylene-vinyl acetate copolymer and wax, and, if desired, a tackifier for application in a field requiring tackiness and adhesiveness.

3) Others

Composition composed of ethylene-vinyl acetate and asphalt are used as coating compositions and adhesive compositions for the outer surfaces of plastics pipe made of polyethylene, polyvinylchloride, ABS, etc., or metal pipe made of steel, copper, aluminium, etc.; and as lining agents or bonding agents for building materials; and as heat insulation for automobiles and buildings.

A tackifier can be added for imparting tackiness and adhesive properties, and a plasticizer, a rubber or a filler can also be added. The compositions composed of ethylene-vinyl acetate copolymer, a tackifier, a pigment, a plasticizer and a filler can be used as hot melt traffic paints for coating a colored composition onto the surface of roads, or as hot melt paints.

The characteristics required for the hot melt compositions are high adhesion to the substrate, softness in the solid state, and excellent heat resistance (high softening tem-

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perature) and cold resistance. However, the softening temperatures of hot melt compositions are usually lower than 100°C and thus are not high enough to prevent separation from the substrate in hot water or in other heated conditions. Also, it is necessary that the compositions have a low melt viscosity for ease of processing, even though a high softening temperature is required.

10. In conventional hot melt compositions, in order to improve processability and wettability and to decrease the melt viscosity of the hot melt compositions, materials having as a main component an ethylene-vinyl acetate copolymer having a high melt index and high vinyl acetate content have been used. Relatively large amounts of paraffin wax (melting point 100—250°F), coal wax (melting point 90—115°C), low molecular weight polyethylene wax (melting point 80—110°C), plasticizers, or the like have been added, so that the content of ethylene-vinyl acetate copolymer is decreased. However, paraffin wax, coal wax, low molecular weight polyethylene waxes, etc. are hard and fragile and hot melt compositions containing these components are deficient in heat resistance (softening temperature), softness, adhesion and cold resistance. When a large amount of paraffin wax or plasticizer is added, the heat resistance (softening temperature) is even further decreased. The melt viscosity of the hot melt composition can be decreased by increasing the melt index or vinyl acetate content or decreasing the content of ethylene-vinyl acetate copolymer.

35. The tensile strength and melting point of the ethylene-vinyl acetate copolymer will decrease with increasing melt index and vinyl acetate content. Accordingly, when a hot melt composition is prepared using an ethylene-vinyl acetate copolymer having a high melt index and high vinyl acetate content, the adhesion (cohesion) and the heat resistance of the composition will decrease. When the content of the ethylene-vinyl acetate copolymer is decreased, the adhesion properties, softness and cold resistance of the composition will decrease.

50. As stated above, the adhesion, heat resistance (softening temperature) softness and cold resistance of the hot melt compositions will substantially decrease by decreasing the melt viscosity of the compositions in attempting to improve processability. In order to increase the heat resistance (softening temperature) of the hot melt compositions, ethylene-vinyl acetate copolymers having a low content of vinyl acetate, high melting point additives or fillers have been used. The heat resistance of the hot melt composition is increased by these operations, but the softness and the cold resistance are decreased and the melt viscosity is increased. It has thus been difficult to obtain a hot melt composition having a low melt viscosity, high adhesion, softness and cold

resistance at the same time as high heat resistance (high softening temperature).

We have studied various methods of improving the heat resistance of hot melt compositions without causing deterioration in other important properties of the hot melt compositions; especially various methods of increasing the softening point to higher than 100°C.

We have found that this can be attained by blending in the composition a modifier of an oxidized low molecular weight propylene-ethylene copolymer, having a molecular weight of 500—10000 and an acid value of 1—20 and an ethylene content of 1—20 wt. %, preferably 1—15 wt. %, the weight ratio of modifier to content of ethylene-vinyl acetate copolymer and/or rubber being 1/100—60/10.

In the hot melt compositions of the invention, the main component is a rubber and/or an ethylene-vinyl acetate copolymer having a melt index of 1—1000 g/10 min. and a vinyl acetate content of 10—45 weight percent. The rubber can be selected from various rubber-like materials and preferably has a Mooney viscosity (25°C) of 1—20, especially styrene-butadiene block copolymers such as styrene-butadiene or styrene-isoprene block copolymers (Encyclopedia of Polymer Science and Technology especially Vol. 15, P. 508—530) (styrene content 5—50%, preferably 20—40% by weight). The composition is solid at room temperature.

Preferably, the compositions to which the modifier is added are composed of

(1) 1—60 weight, more preferably 10—40 weight percent of the ethylene-vinyl acetate copolymer and/or a rubber, and

(2) at least one of the following: 10—70 weight percent, preferably 10—50 weight percent of a tackifier; 1—80 weight percent, preferably 20—70 weight percent of a wax; and 10—90 weight percent, preferably 40—90 weight percent of an asphalt; and

(3) if desirable, one or more of the following: 1—20 weight percent, preferably 1—15 weight percent of a plasticizer; 1—30 weight percent, preferably 1—20 weight percent of a non-tacky resin; 1—60 weight percent, preferably 1—20 weight percent of a filler; and a pigment. If rubber is included in addition to the ethylene-vinyl acetate copolymer, it is preferably in an amount of 1—20 weight percent, more preferably 1—15 weight percent.

Preferred compositions to which the modifier is added are as follows:

	weight percent	
(1) ethylene-vinyl acetate copolymer	5—60	125
tackifier	10—70	
wax	15—30	

		weight percent	indene resins and styrene resins. The petroleum resins may be aliphatic petroleum resins, alicyclic petroleum resins or aromatic petroleum resins. The styrene-related resins can be styrene resins and substituted styrene resins. Suitable petroleum resins may be prepared from thermally cracked residual oil and are solid at room temperature. They impart tackiness and adhesion to the hot melt composition. Aliphatic petroleum resins can be prepared mainly by polymerization of C ₄ and C ₅ fractions which include butene, isobutylene, 1,3-pentadiene-cyclopentadiene or the like as main components. Aromatic petroleum resins can be formed from fractions containing alkylstyrenes, e.g. α -methylstyrene, vinylstyrene; indene and alkylindene as main components. Alicyclic petroleum resins can be prepared mainly by polymerization of C ₄ and C ₅ fractions.	65
	(2) ethylene-vinyl acetate copolymer	25—60		
5	tackifier	20—70		
	filler	5—50		
	(3) ethylene-vinyl acetate copolymer	5—60		70
	tackifier	20—70		
10	plasticizer	5—15		
	(4) ethylene-vinyl acetate copolymer	5—60		75
	tackifier	20—50		
	wax	20—30		
15	non-tacky resin	5—20		
	(5) ethylene-vinyl acetate copolymer	20—40		80
	wax	1—80		
20	(6) styrene-butadiene block copolymer	5—60		
	tackifier	10—70		

Compositions (1) and (2) are especially preferable.

- 25 Suitable tackifiers should be miscible with the ethylene-vinyl acetate copolymer and/or rubber, and are usually selected from natural products, derivatives of natural products and synthetic resins. Suitable natural products and their derivatives include compounds having polar groups, such as rosins, natural resins, terpenephennol resins; compounds having no polar groups, such as terpene resins; α -pinene or β -pinene-related terpene resins and dipentene resins. The rosin resins have, as a main component, abietic acid and abietic esters. Suitable rosins include gum rosins obtained from pine trees; wood rosins obtained by solvent extraction of the roots of pine trees and tall oil rosins obtained from pulp waste.
- 30 Suitable rosins also include modified rosins, hydrogenated rosins, disproportionated rosins, and polymerized rosins. Suitable rosin derivatives include the polyol derivatives of rosins, hydrogenated rosins, polymerized rosins and disproportionated rosins. Suitable polyols include methanol, triethyleneglycol, diethyleneglycol, ethyleneglycol, glycerol and pentaerythritol.

- 50 Suitable natural resins having a polar group include damar, copal, shellac and terpenephennol. Natural resins having no polar group include terepene resins, α -pinene-related terpene resins, β -pinene-related terpene resins, and diterpene resins. The terpene resins can be obtained from turpentine. The terpenes are polymerized to resins of varying polymerization degree, molecular weight, distribution of molecular weight and copolymerization, which are in the form of viscous liquids or solids.
- 55 Any terepene resin is suitable.

- 60 Suitable synthetic resin tackifiers (including hydrocarbon resins) include polymerized resins and polycondensed resins. The polymerized resins may be petroleum resins, coumarone-

The coumarone-indene polymerized resins are composed of a coumarone-indene main component of a residual oil. Various copolymers of coumarone-indene are also suitable. Coumarone-indene-styrene and related resins are commercially available. The polymerized styrene resins can be low molecular weight styrene resins, substituted styrene resins, styrene copolymers, or the like. Low molecular weight styrene resins, α -methylstyrene-vinyl toluene copolymer and related compounds are commercially available.

Suitable synthetic resin and polycondensed resin tackifiers include phenol resins, alkylphenol resins, rosin modified phenol resins and xylene resins. Tackifiers suitable for use are resins which are solid at room temperature, are miscible with the ethylene-vinyl acetate copolymer and rubber and have a melting point of 40—180°C.

Suitable waxes include animal wax, botanical (phyto) wax, mineral wax and synthetic wax. The animal wax can be beeswax, spermacetic wax, wool fat, shellac or the like. The botanical wax can be caruba wax, candelilla wax, esparto wax, ouricoury wax, etc. The mineral wax can be natural wax, petroleum wax, and others. The petroleum wax can be paraffin wax, motor oil wax or micro wax. The synthetic wax can be polyethyleneglycol wax, blend terphenyl, halogenated hydrocarbons, Fisher-Tropsch polyethylene wax, hydrogenated wax, or the like. Petroleum wax is preferably used for hot melt compositions, especially hot melt coating compositions and includes crystalline and micro-crystalline wax. Suitable crystalline waxes include 100 paraffin to 145 paraffin, as classified by melting point in the Japanese Industrial Standard K-2235-1961. The 135 paraffin and 145 paraffins are used mainly for hot melt coating, and they have good miscibility with ethylene-vinyl acetate copolymer. The microcrystalline waxes are classified in grades having melting points higher than 155°F, higher than 165°F, 170—

175°F, 180—190°F, 190—197°F and 197—220°F, by ASTM. The grade having melting points of 170—175°F is preferable. The wax is blended with the ethylene-vinyl acetate copolymer, and if necessary, with a tackifier to impart tackiness, and a desirable other component. The composition is then coated onto a substrate.

The coated products should have excellent antiblocking properties, lustre, moisture permeability and flexibility, as well as excellent heat resistance, cold resistance, and adhesion to substrate. The heat resistance, cold resistance and adhesion to substrate are indispensable.

Suitable asphalts include blown asphalt and strate asphalt having softening points of 40—130°C, measured by the method of Japanese Industrial Standard K-2531-60, and needle penetration values less than 100, measured by the method of Japanese Industrial Standard K-2530-60. The composition of the asphalt and ethylene-vinyl acetate copolymer plus any other necessary component is solid.

Pigments may be added to make hot melt adhesive compositions, hot melt coating compositions, traffic paints, hot melt paints or the like. Most pigments, such as inorganic pigments, used in conventional traffic paints and hot melt paints are suitable. Suitable pigments include titanium oxide, zinc sulfide, lithopone, basic lead sulfate, basic lead carbonate (white lead) and zinc oxide as white pigments; carbon black and iron black as black pigments; cadmium red, vermilion, molybdenum red and red lead as red pigments; red oxide, amber and sienna as brown pigments; yellow lead, cadmium yellow, zinc yellow and ocker as yellow pigments; chromium green, chromium oxide and zinc green as green pigments; ultramarine, Berlin blue and cobalt blue as blue pigments; and cobalt violet and manganese violet as violet pigments.

Fillers may be dispersed in the hot melt composition, and impart antiblocking, threading and other properties. Fillers used in conventional hot melt compositions, traffic paints, and hot melt paints are suitable, and include barium sulfate, calcium carbonate, magnesium carbonate, titanium oxide, hydrated silica and magnesium oxide, which have a density of about 1.0—5.0 g./cc.

Rubber may be added to the hot melt composition mainly to impart high cold resistance and danti-softness. Suitable rubbers include polyisobutylene, butyl rubber, styrene-butadiene rubber, acrylonitrile-butadiene rubber, 2-chlorobutadiene-1, 3 polymer chloroprene rubber, polybutadiene and ethylenepropylene rubber. The rubber is added to the extent of 0.5—20 weight percent to the hot melt type composition.

Plasticizers may be added to the hot melt composition, mainly to impart flexibility. Suitable plasticizers include phthalic esters, such

as 2-ethylhexyl phthalate, dimethylphthalate, diethylphthalate, dibutylphthalate, diheptyl phthalate, 2-diisooctyl phthalate, di-n-octyl phthalate, 2-dinonyl phthalate, diisodecyl phthalate, dodecyl phthalate, dicyclohexylphthalate and dilauryl phthalate; dibasic aliphatic acid esters, such as dioctyl adipate and diisodecyl adipate; aliphatic acid esters such as butyl oleate and methyl acetyl ricinolate; phosphoric acid esters, such as tricresyl phosphate, trioctyl phosphate and triphenyl phosphate; polymers, e.g. polyesters; paraffins, e.g. chlorinated paraffin; trioctyl trimellitate, and others. The plasticizer is preferably added in amounts less than 40 weight percent to the hot melt composition to maintain miscibility with the other components and prevent blooming.

Other polymers may be added to impart suitable characteristics, such as improved tensile strength and mechanical properties, which may be desired depending upon the intended applications. Suitable polymers include atactic polypropylene, polyethylene, ethylene-organic acid copolymer, oxidized polyethylene and low molecular weight ethylene-vinyl acetate copolymer, and others.

Suitable hot melt compositions of this invention include hot melt adhesive compositions, hot melt coating compositions, asphalt blends traffic paints and hot melt paints, which are prepared by blending the above components, depending upon the intended application. The components are heated and mixed to prepare the compositions. A tank equipped with a stirrer is preferably used as a means for heating, melting and mixing, but any type of apparatus, which can heat and mix the contents, can be used. An Intensive mixer, Banbury mixer, or Super mixer is preferable.

To prepare the hot melt composition in the form of a pellet, rod or thread, the composition is heat-melted, mixed in an extruder equipped with a screw, extruded into a mold cooled to form the desired shape. The operation of heating, melting and mixing is preferably performed under atmospheric pressure but may be performed under high pressure or reduced pressure. The temperature for heating, melting and mixing is dependent on the component having the highest melting point, and is usually in the range of 70—300°C, preferably 90—200°C.

In the preparation of the hot melt composition by the process of this invention, the heat resistance (softening temperature) of the composition is improved by adding an oxidized low molecular weight propylene-ethylene copolymer as modified having an acid value of 1—20 per molecule, and an ethylene content of 1—20 wt. %, preferably 1—15 wt. %, the weight ratio of modifier to content of ethylene-vinyl acetate copolymer and/or rubber being 1/100 to 60/10.

The oxidized low molecular weight propylene-ethylene copolymer is prepared by oxidiz-

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ing low molecular weight propylene-ethylene copolymer with oxygen or an oxygen-containing gas or ozone. When steam is supplied in the oxidation step with oxygen, the oxygen-containing gas or ozone, oxidized low molecular weight propylene-ethylene copolymer having excellent color can be obtained. The oxidized low molecular weight propylene-ethylene copolymers used in the invention have an average molecular weight of 500—10,000 (measured by intrinsic viscosity) and an acid value of 1—20 per molecule and an ethylene content of 1—20 wt. % preferably 1—15 wt. %.

We have found that, when using the teachings of this invention, the softening temperature of the resulting composition (measured by the ring-ball method) can be raised to higher than 100°C and melt viscosities at 160°C (measured by Brookfield viscometer) of 6000 cps or less. The melt viscosity of conventional hot melt compositions at 180°C is more than 10,000 cps.

Having generally described the invention, a further understanding can be obtained by reference to the following specific examples which are provided herein for purpose of illustration only and are not intended to be limiting in any manner.

Examples

The following components (weight %) were heat-melted and mixed at 120—200°C for 1—2 hours, and the physical properties of the resulting hot melt composition were tested. In the Examples, the following abbreviations are used.

EVA—ethylene-vinyl acetate copolymer

VA—vinyl acetate

MI—melt index

MW—molecular weight (measured by intrinsic viscosity)

Example 1

EVA (VA content 28%, MI 400)	40%
Glycerine ester of hydrogenated rosin (R & B softening temperature 72°C)	45%
Oxidized low molecular weight propylene-ethylene copolymer (MW 3500; Acid value 5) (ethylene content 8%)	15%

Properties of the composition

R & B softening temperature	125°C
Melt viscosity (160°C)	2700 cps
Peel strength (A1/A1)	11.0 kg/25 mm
Tensile strength	15.0 kg/cm ²
Elongation	1800%
Hardness (Shore-A)	45
Cold resistance	Good
Heat seal strength (OPP/OPP)	3.0 kg/15 mm

Note:

Test methods

- (1) R & B softening temperature Japanese Industrial Standard K 2531 65
- (2) Melt Viscosity Brookfield viscometer
- (3) Peel strength: 90° peeling Substrate: 25 mm width 0.1 mm thickness aluminium foil 70
- Structure: Al foil/hot melt/Al foil comp. (0.2 mm thickness)
- Peeling speed 150 mm/min. 75
- (4) Tensile strength and Elongation ASTM D 638—68
- (5) Hardness: ASTM D 2240—68
- (6) Cold resistance: A hot melt composition which was melted, was coated on a corrugated board and another corrugated board was plied onto the coated surface under pressure. The product was kept at -30°C for 3 hours and the product was peeled after warming to room temperature. When the corrugated board was broken, the cold resistance was good. When the coated surface broken, the cold resistance was bad. 80
- (7) Heat seal strength: The hot melt composition was coated on OPP in a thickness of 20 μ and another OPP was plied onto the coated surface. The plies were heat-sealed at 80°C under a pressure of 1 kg/cm² for 1 second. The product was peeled at a peeling speed of 200 mm/min. 85

Example 2

EVA (VC content 19% MI 350)	40%	100
Hydrogenated dicyclopentadiene resin (R & B softening temperature 123°C)	30%	
Oxidized low molecular weight propylene-ethylene copolymer (MW 4900 acid value 15) (ethylene content 5 wt.%)	30%	105

Properties of the Composition

R & B softening temperature	135°C	110
Melt viscosity (160°C)	3500 cps	
Peeling strength (Al/Al)	6.5 kg/25 mm	
Tensile strength	25 kg/cm ²	
Elongation	900%	115
Hardness (Shore-A)	50	
Cold resistance	Good	

Example 3

EVA (VA content 28% MI 400)	40%	120
Hydrogenated rosin (R & B softening temperature 70°C)	40%	

	Oxidized low molecular weight propylene-ethylene copolymer (MW 4000 acid value 3.5) (ethylene content 10 wt. %)	20%	Peel strength (Al/Al)	3.5 kg/25 mm	45
			Cold resistance	Good	
5	Properties of the Composition		Example 5		
	R & B softening temperature	138°C	EVA (VA content 28%, MI 400)	40%	50
	Melt viscosity (160°C)	5500	Polymerized rosin	20%	
10	Peeling strength (Al/Al)	10 kg/25 mm	Oxidized low molecular weight propylene-ethylene copolymer (MW 2000 Acid value 3.5) (ethylene content 10 wt. %)	40%	55
	Tensile strength	35 kg/cm ²	R & B softening temperature	140°C	
	Elongation	1200%	Melt viscosity	4500 cps	
	Hardness (Shore—A)	80	Peel strength (Al/Al)	3.0 kg/25 mm	60
	Cold resistance	Good	Elongation	500%	
15	Heat seal strength	3.5 kg/15 mm	Example 6		
	As a reference, low molecular weight poly-ethylene (MW 1500, R & B 102°C) was substituted for the oxidized low molecular weight propylene-ethylene copolymer.		EVA (VA content 27%, MI 150)	20%	65
20	The properties of the hot melt compositions were as follows. The heat resistance, adhesiveness, softness and cold resistance of the compositions were quite inferior to those of the former composition.		Wax (softening temperature 145°F)	60%	
	R & B softening temperature	86°C	Oxidized low molecular weight propylene-ethylene copolymer (MW 2000 Acid value 3.5) (ethylene content 3 wt. %)	20%	70
25	Melt viscosity	2800 cps	R & B softening temperature	120°C	
	Peel strength	1.5 kg/25 mm	Melt viscosity (160°C)	1000 cps	
	Tensile strength	20 kg/cm ²	Peel strength (Al/Al)	1.5 kg/25 mm	
	Elongation	350%	As a reference, the properties of the composition of EVA (no oxidized low molecular weight propylene-ethylene copolymer) were tested. The cold resistance, moisture permeability and peel strength of the composition were inferior.		75
30	Hardness (Shore—A)	79	EVA (VA content 27% MI 150)	30%	80
	Cold resistance	Bad	Wax (softening temperature 145°F)	70%	
	Heat seal strength (OPP/OPP)	0.2 kg/15 mm	R & B softening temperature	70°C	85
	Example 4		Melt viscosity (120°C)	640 cps	
35	EVA (VA content 28% MI 400)	40%	Peel strength (Al/Al)	120 g/25 mm	
	Cyclopentadiene resin	30%	Moisture permeability	9 g/m ² . 24 Hr.	
40	Oxidized low molecular weight propylene-ethylene copolymer (MW 2000 Acid value 3.5) (ethylene content 15 wt. %)	30%	Example 7		
	R & B softening temperature	118°C			
	Melt viscosity	3000 cps			
90	Styrene-butadiene block copolymer (styrene content 30% Mooney viscosity 10)		①	②	
	Polymerized rosin		40%	40%	
95	Hydrogenated rosin (R & B softening temperature 70°C)		—	20%	
	Oxidized lower molecular weight propylene-ethylene copolymer (MW 4000 acid value 3.5) (ethylene content 10 wt. %)		40%	—	
100	Properties of the Composition		20%	40%	
	R & B softening temperature				
	Melt viscosity (160°C)		136°C	140°C	
	Peeling strength (Al/Al)		16500	13500	
105	Elongation		9.5 Kg/25 mm	2.8 Kg/25 mm	
			—	500%	

WHAT WE CLAIM IS:—

1. A hot melt composition which comprises an ethylene-vinyl acetate copolymer, having a vinyl acetate content of 10—45 weight percent and melt index of 1—1000 g/10 min., and/or a rubber, the composition having admixed therewith an oxidized low molecular weight propylene-ethylene copolymer as modifier having an acid value of 1—20, and an ethylene content of 1—20 wt. % and having an average molecular weight of 500—10,000, the weight ratio of modifier to content of ethylene-vinyl acetate copolymer and/or rubber being 1/100—60/10.
2. The hot melt composition of claim 1 comprising at least one additive selected from a tackifier, wax and asphalt.
3. The hot melt composition of claim 2, wherein the tackifier is a rosin, modified rosin or petroleum resin.
4. The hot melt type composition of claim 2 or 3, wherein the wax is a crystalline wax or microcrystalline wax.
5. The hot melt composition of any one of claims 1 to 4 wherein the rubber has a mooney viscosity (25°C) of 1—20.
6. The hot melt type composition of claim 1, wherein the rubber is styrene-diene block copolymer having a mooney viscosity (25°C) of 1—20 and a styrene content of 5—50 wt. %.
7. The hot melt composition of any one of claims 1 to 6 wherein the ethylene content of the modifier is 1—15 wt. %.
8. The hot melt composition of any one of the preceding claims which has an R & B softening temperature of greater than 100°C and a melt viscosity of not more than 6000 cps at 160°C.
9. The hot melt composition of any one of the preceding claims wherein 1—60 weight percent of the ethylene-vinyl acetate copolymer and/or rubber is employed, and at least one additive selected from
 - 10—70 weight percent of tackifier;
 - 1—80 weight percent of wax; and
 - 10—90 weight percent of asphalt.
10. The hot melt composition of claim 9, wherein 10—40 weight percent of the ethylene-vinyl acetate copolymer and/or rubber is employed, and at least one additive selected from
 - 10—50 weight percent of tackifier;
 - 20—70 weight percent of wax; and
 - 40—90 weight percent of asphalt.
11. A hot melt composition according to claim 1 which includes the ingredients as specified in any one of compositions (1) to (6) described herein in addition to the modifier.
12. A hot melt composition according to claim 1 substantially as herein described with reference to any one of Examples 1 to 7.
13. A method of raising the softening temperature of a hot melt composition which comprises: blending an oxidized low molecular weight propylene-ethylene copolymer having a molecular weight of 500—10000, an acid value of 1—20 and an ethylene content of 1—20 wt. % in a weight ratio of 1/100—60/10 with respect to the content of an ethylene-vinyl acetate copolymer, having a vinyl acetate content of 10—45 wt. % and a melt index of 1—1000 g/10 min., and/or a rubber having a mooney viscosity (25°C) of 1—20 with which are further blended at least one additive selected from a tackifier, wax and asphalt.
14. A method according to claim 13 substantially as herein described with reference to any one of Examples 1 to 7.
15. A hot melt composition obtained by the method of claim 13 or 14.
16. Articles having a coating of or employing as an adhesive a hot melt composition according to any one of claims 1 to 12 or 15.

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